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A MODEL FOR DISPERSION FROM AREA SOURCES IN CONVECTIVE TURBULENCE

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Abstract—Vertical mixing coefficients have been computed by integrating vertically changes of concentrations of nonreactive pollutants along horizontal trajectories, during convective conditions.

Mixing coefficients are obtained for three separate periods, and analyzed according to the hypothesis of convective similarity. It was found that normalized mixing coefficients could be represented as "universal" functions of the ratio of the height to the mixing depth. These functions were small at small z and large z and reach a maximum at about half the mixing depth. In fact, the K-coefficients are so large in the middle of the boundary layer, that the concentrations there are effectively independent of height.

In the surface layer, the mixing coefficients agree with the hypothesis that mixing coefficients for contaminants equal mixing coefficients for momentum (eddy viscosity). The observed universal functions also agreed fairly well with predictions made by Lumley and Zeman from second-order closure theory. However, laboratory measurements indicate larger mixing coefficients. It is suggested that K-values estimated both from second-order closure theory and from Los Angeles measurements are systematically underestimated. Nevertheless, it seems likely that K-theory is useful for determining pollutant concentrations from large, continuous area sources at the ground, under convective conditions.

1. INTRODUCTION

In the Fall of 1973, a large, cooperative pollution project was carried out at Los Angeles, under the general direction of William Perkins. The project is usually referred to as LARPP, for Los Angeles Reactive Pollution Project. Many organizations cooperated, private and federal. Most of the funding was provided by the Coordinating Research Council of New York.

The emphasis in this project was on the study of reactive contaminants; but, as will be seen, the behavior of stable contaminants as measured during LARPP gives valuable information about dispersive properties of the atmosphere. Without an understanding of such properties, the characteristics of reactive contaminants cannot be understood satisfactorily.

Based on measurements during just one operation (operation 33), one of the authors carried out a pilot project for determining the diffusion characteristics for Los Angeles on sunny mornings. This project was made possible by the fact that concentration measurements were made in a Lagrangian framework; that is, along air trajectories. A report on these early results was published by Panofsky (1975). This paper did not show to what extent the conclusions could be generalized to other conditions. Additional operations of the LARPP project have now been analyzed and a technique is suggested for the generalization of the results on the basis of the assumption of convective similarity.

2. THEORY

Concentrations of reactive contaminants over cities are often described by diffusion equations of the form:

$$\frac{d\chi}{dt} = \frac{\partial}{\partial z} K \frac{\partial \chi}{\partial z} + \text{Sources} - \text{Sinks}. \tag{1}$$

Here, χ is the concentration of a contaminant in mass per unit volume, t is time, z is height and K the vertical diffusion coefficient. It is assumed that horizontal diffusion and vertical advection can be neglected. The lefthand side of Eq. (1) expresses the change of concentration along a smoothed horizontal air trajectory.

In order to apply Eq. (1) properly to reactive contaminants, the characteristics of K must be known. K can be estimated from observations of concentrations of stable contaminants made during LARPP, as will be shown.

In general, the equation of continuity of a nonreactive contaminant is assumed to be of the form:

$$\frac{d\chi}{dt} = -\frac{\partial F_z}{\partial z} \tag{2}$$

where $d\chi/dt$ is the change of χ with the time along a trajectory. F_z is the vertical flux of the contaminant at level z. Again, advection by vertical motion and horizontal diffusion are neglected.

Early in the morning, observations are available up to the height of the mixed layer, h. In that case, F_z is computed from

$$F_z = \int_z^h \frac{d\chi}{dt}\,dz. \tag{3}$$

Equation (3) yields fluxes at various levels, including at the ground. LARPP data were collected from helicopters in such a way as to make computations of $d\chi/dt$ possible. The mean height of the lowest inversion base, z_i, is typically 10–15% less than the maximum height reached by the pollutants and is often used in its place.

After the top of the mixed layer passes beyond the largest height of observations (about 250 m), F_z is computed from

$$F_z = F_0 - \int_0^z \frac{d\chi}{dt}\,dz. \tag{4}$$

F_0, the surface flux is obtained from the earlier computed surface fluxes by applying a factor depending on the diurnal variation of traffic, because the contaminants studied (except CH_4) originate primarily from automobiles. The factor was derived from traffic counts published by Nordsieck (1974). The surface flux of methane is put equal to zero.

Given F_z, K is computed from its definition:

$$K = -\frac{F_z}{\dfrac{\partial\chi}{\partial z}}. \tag{5}$$

This technique for obtaining K was first used by McCormick (unpublished). In general, $\partial\chi/\partial z$ could be estimated from slopes of graphs of χ plotted as function of height. Near the ground, the slope changes rapidly with height, and concentrations were only given at the surface and at 75 m. Therefore, $\partial\chi/\partial z$ at 50 m was estimated from values of F_0, and from χ at the ground and at 75 m. Since the wind was weak and the turbulence was convective, it was assumed that $\partial\chi/\partial z$ varies as $z^{-4/3}$. If we then take the effective level of the "surface" observations as 10 m, $(\partial\chi/\partial z)$ at 50 m is almost exactly $(\chi_{75}-\chi_{10})/150$ m, where subscripts refer to heights of observations.

The surface heat flux H was obtained from an equation analogous to Eq. (3):

$$H = c_p\rho \int_0^h \frac{dT}{dt}\,dz. \tag{6}$$

Here ρ is the air density, T temperature and c_p the specific heat at constant pressure.

In order to generalize results from one experiment to another, it is often possible to introduce a similarity theory appropriate for the situation. Once the variables have been properly scaled, this type of theory then reduces to the problem of evaluating functions which are presumably universal, and are usually obtained from observations; occasionally the form of such functions can also be predicted theoretically.

For the Los Angeles situation on sunny days with weak winds, "convective scaling" seems appropriate. This is true because the Monin–Obukhov scale has a magnitude of only a few meters so that $h/|L|$ is much larger than 10. For that purpose it is the custom to introduce a length scale, h and a velocity scale, w_*. The length scale h is here taken as the mixing depth, but is more commonly taken as the height of the lowest inversion, z_i. This can either be inferred from observations, or computed, given the vertical heat flux at the ground, H, the sounding before sunrise, and the large-scale vertical velocity. The theory has been discussed, for example, by Tennekes (1973). The scaling velocity w_* is defined by:

$$w_* = \left(\frac{gHh}{c_p\rho T}\right)^{1/3}. \tag{7}$$

Here, T is temperature, ρ density, g gravity and c_p the specific heat at constant pressure.

In terms of these scaling quantities the mixing coefficients should follow the relationship

$$K_* = \frac{K}{w_* h} = f\left(\frac{z}{h}\right) \tag{8}$$

where $f(z/h)$ is the vertical distribution function for K. The function $f(z/h)$ can be assumed to be universal under the conditions that (1) the turbulence structure within the mixed layer is self-similar and in equilibrium with current boundary conditions, and (2) the normalized eddy coefficient K_* is independent of the nature of the pollutant source distribution. The condition (1) is usually met in practice; it requires that the mixed layer vary slowly ($\partial h/\partial t \ll w_*$). Also, according to Deardorff and Willis (1976), self-similarity occurs only after $w_* x/uh$ exceeds 2.5 (where x is distance covered after the pollutant has been inserted and u is the mean wind speed). This condition is also satisfied. The condition (2) is, in general, not satisfied in buoyant flows; however, we can compare K_* for situations with similar source distributions. In this paper we deal with large surface sources which, to a first approximation, can be taken as horizontally homogeneous infinite area sources. Of course, the source strength changes with time. But, as will be seen, there does not appear any systematic change of $f(z/h)$ with time. Hence, the premise of universality of the function (z/h) for the situations encountered in this paper may be justified.

Once f is known, K can be derived generally under convective conditions, given estimates of heat flux and of h. On sunny, windy days, a correction can easily be made for the additional diffusion in the surface layer by mechanical turbulence. It is the purpose of this note to describe the derivation of $f(z/h)$ from LARPP data, and to make some tests of its generality.

3. THE OBSERVATIONS

Concentrations of many contaminants were determined from helicopters every 6 s. The helicopters followed smoothed horizontal air trajectories determined from the centers of gravity of three balloons (constant-level balloons, tracked by radar). The helicopters would fly around squares about 2 miles on the side, at fixed levels, before proceeding to another level. Many other data were obtained including ground concentrations, surface winds and radiation data. Unfortunately, upper winds and temperatures were measured at two locations only, and only about sunrise and near noon.

Altogether, 46 operations were carried out during LARPP, from Sept 11–Nov. 20, 1973. A tape containing all measurements during these operations was kindly made available by A. Eschenroeder.

In order to make the computations implied by Eqs. (3) through (6) legitimate, the following conditions had to be satisfied by the observations:

(a) The data gathering process had to have started sufficiently early so that h did not penetrate the largest height of measurement before about the first hour of observation; this condition could be relaxed if h never exceeded 400 m or so, as might be the case when strong subsidence occurred.

(b) The trajectories of the balloons and helicopters did not traverse regions with hills of significant heights.

(c) The observation density from 50 m–300 m was great enough to allow reliable empirical determinations of Lagrangian derivatives $d\chi/dt$.

In only three operations all these conditions were satisfied, those numbered 23, 31 and 33. Of these, operation 33 had been studied previously; but the period of analysis for this operation could be extended beyond the original period by use of Eq. (4).

Of course not all the observations taken during the three operations were required. The following information was analyzed.

(a) Graphs were constructed of differences between height above sea level and height above surface, thus describing terrain variations. If the terrain varied significantly, the operations were not analyzed further.

(b) Concentration of passive contaminants were obtained along the air trajectories during the first 2h or so. Thus, concentrations of CO, CH_4 and $NO + NO_2$ were used, even though there is some controversy whether the sum of nitric oxide and nitrogen dioxide concentrations is sufficiently conservative for this purpose. The actual concentrations analyzed represented averages over at least 10 consecutive observations. In more than half of the cases, the averages represented 50 individual observations. If the standard deviations of the individual observations about the mean was half the mean or larger (in about 1% of the cases), the average was discarded. In all cases means were computed only if the heights above sea level as well as the heights above terrain had ranges of less than 50 m.

(c) Soundings near sunrise and noon at El Monte were used in the determination of mixing depth.

(d) Surface heat flux was inferred from temperatures as observed along the trajectories from the helicopters.

(e) Insolation was obtained from James Peterson, EPA, to compare with surface heat flux.

4. ANALYSIS OF THE OBSERVATIONS

First, diagrams were plotted with concentration of stable contaminant as ordinate and time as abscissa. Heights of the helicopter were written into the diagrams, and smoothed isopleths of height were estimated. These lines represent the variation of contaminants with time (along a horizontal trajectory at a given height). Figure 1 gives an example.

All figures have the same general behavior. Early in the morning the concentrations above the surface are low. As the inversion lifts, and the mixed layer rises beyond a given height, the concentrations go up at that height. As the mixed layer grows further and the strength of the ground sources diminishes, the concentrations decrease. The pollutants reach the low levels early, and the peaks are strongest at low levels, so that there is generally a decrease of concentrations upward. Also, peaks occur later at the higher levels. Late in the morning, the vertical gradient becomes too small to measure. At that time, the observed region of the boundary layer becomes "perfectly mixed"; $\partial\chi/\partial z$ approaches zero. Eq. (5) then shows that K essentially becomes infinite; at least, it is too large for measurement.

Figures such as Fig. 1 can be used to establish h at various times in the early morning—a very important fact—otherwise the mixing depth is only given near noon, from the soundings at that time. The time just before concentrations begin to rise is the time at which $z = h$. Thus, Fig. 2 shows the inferred progression of h with time for the three periods analyzed. Since relatively few points are available to draw the lines on this graph, straight lines were used to represent the variation of h with time.

Graphs such as Fig. 1 were used directly to determine $d\chi/dt$ at 15-min intervals. Then, $d\chi/dt$ was plotted as function of height, assuming that this quantity

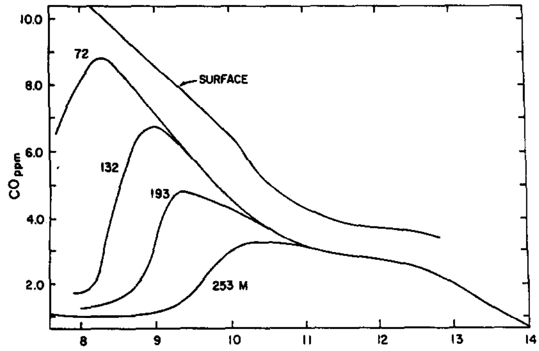

Fig. 1. Distribution of CO concentrations as functions of time and height, operation 33.

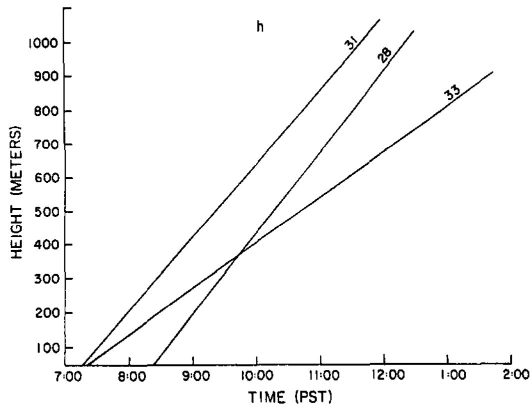

Fig. 2. Growth of thickness of mixed layer h for operations 28, 31 and 33.

goes to zero at $z = h$. Finally, areas under these curves between arbitrary height z and either h (for the first hour) or between z and the ground (later) were estimated to evaluate the vertical fluxes according to Eqs. (3) or (4). Since there are relatively few points on these diagrams, the fluxes determined in this manner are subject to considerable uncertainty. This is especially true for the surface fluxes in the first hour, which are composed of sums of negative and positive contributions.

Next, χ-values were taken from graphs such as Fig. 1 and plotted as function of height. Smooth lines were drawn through these points, to represent local concentration profiles. But, because of the relatively few points, the profiles, and particularly their gradients, are quite uncertain. Nevertheless, K-values were obtained from the computed fluxes and concentration profiles according to Eq. (5). As was true for operation 33, the other runs also show that roughly the region $0.2 < z/h < 0.55$ develops vertically uniform distributions of pollutants in the Los Angeles boundary layer, so that K-values there are too large to determine.

5. SYNTHESIS

In order to obtain a general method for the estimation of K in growing convective boundary layers, the results were combined and used as shown in Eq. (8), based on the assumption of convective similarity.

In order to compute K_*, it is necessary first to evaluate the convective scaling velocity, w_*. This quantity depends on the surface heat flux, H (see Eq. (7)). Fortunately, the heat flux occurs only to the 1/3 power, so that great accuracy in its determination is not necessary.

H was determined by evaluating Eq. (6). This proved to be no easy task. Plots of temperature as function of time along the trajectories proved to be erratic. They were smoothed, rather arbitrarily, and derivatives were obtained from the smoothed curves. The vertical integration also proved difficult, especially after the first hour, because of the unknown contribution to the integral of the temperature changes between the highest helicopter observations and h.

Probably, the only really reliable heat flux determinations were made for run 33, during which the temperature variations were relatively smooth. This material was published by Panofsky (1975). It was shown that for period 33, the heat flux was approximately equal to 30% of the insolation an hour earlier. Therefore, insolation measurements were obtained from Dr. James Peterson (EPA) also for operations 28 and 31. From these, and the above relationship found for operation 33, independent heat flux estimates were made. These did not agree well with the values computed from Eq. (6). The final estimates of H were based on Eq. (6) only for operation 31 where the temperature data were reasonably satisfactory; for operation 28 averages were used between the two sets of estimates. Given these heat fluxes, $K_* = K/hw_*$ could then be computed.

Tables 1–4 list the results. For operation 33, the K values were based only on measurement of CO. For operation 28 the values are averages determined from methane and NO_x; and for operation 31, averages for CO and NO_x.

If the similarity hypothesis is correct, the K_* estimates for a given z and a given h should be the same in all operations. This is not quite true; but the differences seem random rather than systematic, a probable consequence of the many uncertainties in the computation of K_*. Further, if K_* depends only on the ratio of z/h, as similarity theory requires, the dependence of K_* on z should be opposite to the dependence on h. Indeed, the table shows that K_* generally decreases with increasing z and decreasing h.

Figure 3 shows the result of plotting K_* as a function of z/h. In the range $0.2 < z/h < 0.55$, $\partial\chi/\partial z$ is effectively zero and K_* too large to measure.

Figure 3 contains solid curves and dashed curves. Basically, the solid curves are drawn to the observations.

The lower solid curve (for $z/h < 0.2$) fits the equation

$$K_M = 2.5\,(kz)^{4/3}\left(\frac{gH}{c_p\rho T}\right)^{1/3} \tag{9}$$

which was suggested by Carl *et al.* (1973) as a representation of the eddy coefficient for momentum transfer under conditions of strong convection in the atmospheric surface layer. (Here k is Von Karman's constant). Thus, the exchange coefficients for contaminants computed here for Los Angeles appear to agree quite well with eddy viscosities.

The dashed curve in Fig. 3 is based on a theoretical model of vertical diffusion from an area source, computed according to the method of second order closure by Zeman and Lumley (1976, 1976a). There are considerable differences between the assumptions in the second-order model and the real world conditions under which the observations were made. In the

Table 1. Fluxes and K-values for operation 28; Flux of NO_x kg m^{-2} s^{-1} × 10^{11}

Height M	Time PST	8:30	8:45	9:00	9:15	9:30	9:45	10:00	10:15	10:30	10:45	11:00	11:15
50		6.3	65	105.1	149.6	43.8	7.5	14.6					
100			25	58.8	98.3	78.8	26.3	38.6					
150				17.5	45.8	72.5	44.8	62.6					
200					13.8	37.5	43.8	76.6					
					1.8	12.5	21.3	65.6					
				Flux of CH_4 kg m^{-2} s^{-1} × 10^9									
50		16.9	63.6	67	30.6	12	12.5	13.5					
100		3.5	29.8	65	46.9	27	30	21					
150			3.2	32.5	45.4	42	47.5	28.5					
200				2.5	21.6	37	60.5	36					
250					0.3	12	62	38					
					K, NO_x m^2 s^{-1}								
50		2.5	6.5	7.0	10.0	12.5	3.0	B*					
100			1.7	4.7	3.0	5.2	10.5	B					
150				1.2	1.3	2.4	2.6	12.5					
200					0.9	1.1	1.3	4.3					
250						0.5	0.7	4.4					
					K, CH_4 m^2 s^{-1}								
50		6.8	8.5	8.9	10.2	12	B	B	B	B			
100		1.4	3.0	7.7	9.4	15	B	B	B	B			
150			0.3	6.5	6.1	10.5	19	B	B	B			
200				0.3	2.2	4.0	9.7	14.4	B	B			
250						0.9	5.5	12.7	B	B			
				Avg. K, CH_4 and NO_x, m^2 s^{-1}									
50		2.5	6.65	7.75	9.45	11.35	7.5	B	B	B			
100			1.55	3.85	5.35	7.2	12.8	B	B	B			
150				0.75	3.9	4.25	6.6	15.8	B	B			
200					0.6	1.65	2.7	9.4	14.4	B			
250						0.3	0.8	5	12.7	B			

* B stands for too big to compute.

model, zero wind is assumed, whereas a slow but distinct wind was observed at Los Angeles; furthermore, the idealized boundary conditions and the assumption of horizontal homogeneity in the model are only a crude approximation to the real situation. In spite of these differences Fig. 3 indicates some agreement between the observed and modeled K_*.

The model experiment permitted experimentation with effects of different source distributions and different growth rates of the mixed layer on the distribution of K_*. The results showed that K_* does vary with the time history of source strength. However when only the initial source history was varied, the later distribution of K_* became independent of the initial variation of source strength.

Also, the model results showed that the rate of growth of h had no influence on the distribution of K_*, provided that

$$\frac{dh}{dt} \ll w_* \tag{10}$$

a condition usually satisfied in atmospheric, convective boundary layers.

The second-order model predictions differ significantly in one particular feature from the observations: in the region in which the observations suggest no variations of χ with height, and hence infinite values of K, the predictions indicate a finite slope of χ and hence a finite K. Plausible reasons for this discrepancy shall be discussed in the next sections.

A further test of the generality of the distribution of K_* is possible on the basis of laboratory measurements by Willis and Deardorff (1976). They observed the dispersion of a contaminant from an instantaneous line source in a box heated from below. By adding concentrations from many hypothetical line sources, they produced a hypothetical distribution of contaminant emitted from an area source at the ground with constant emission rate. The resulting distribution of normalized concentrations χ_* as function of normalized diffusion time $t_* = t\, w_*/z_i$ and of normalized height $\xi = z/z_i$ very nearly satisfies an equation analogous to equation (1):

$$\frac{d\chi_*}{dt_*} = \frac{\partial}{\partial \xi} K_* \frac{\partial \chi_*}{\partial \xi}. \tag{11}$$

In order to compare these measurements with the LARPP results it was assumed that mixing depth exceeded inversion height z_i by 15%.

Table 2. Fluxes and K-values for operation 31; Flux of NO_x, kg m^{-2} s^{-1} $\times 10^{11}$

Height M	Time PST	7:30	7:45	8:00	8:15	8:30	8:45	9:00	9:15
50		50	185	156	122	21			
100			85	121	146	51			
150			5	68	108	73			
200				18	63	78			
250					18	37			
			Flux of CO, kg m^{-2} s^{-2} $\times 10^9$						
50		60	200	402.5	390	111	204	168	
100			65	267.5	400	151	294	228	
150			2.5	120	290	153	360	288	
200				15	155	113	411	338	
250					40	43	446	358	
				K, NO_x, m^2 s^{-1}					
50		3.3	6.2	6.2	12.2	B*			
100			2.6	4.8	8.3	20.4			
150			0.7	2.5	8.6	14.6			
200				0.9	2.6	10.4			
250					0.9	0.9			
				K, CO, m^2 s^{-1}					
50		4	6.7	10.1	15.6	B*	B	B	B
100			1.6	5.4	13.3	30.2	B	B	B
150			0.1	3.4	10.5	10.2	48.9	B	B
200				0.9	3.7	4.2	15.0	B	B
250					0.8	2.9	10.5	11.9	
			K, Avg. CO and NO_x, m^2 s^{-1}						
50		3.7	6.5	8.2	13.9	B	B	B	B
100			2.1	5.1	10.8	25.3	B	B	B
150			0.4	3.0	9.6	12.4	48.9	B	B
200				0.9	3.1	7.5	15	B	B
250					0.9	1.9	10.5	11.9	B

* B stands for too big to compute.

Applying the procedures described by Eqs. (3) through (5), T. Tarbell evaluated K_* in Eq. (11) from the Willis–Deardorff distribution curves. The qualitative distribution of K_* with z/h in this experiment was similar to the LARPP results, with huge values in the central mixed layer. However, level for level, the laboratory K_* were much larger than the Los Angeles values. This difference suggests that the Los Angeles values may have been systematically underestimated. This possibility is discussed in the next section.

Table 3. Fluxes and K-values for operation 33; Flux of CO, kg m^{-2} s^{-1} $\times 10^9$

Height m	Time PST	8:15	8:30	8:45	9:00	9:15	9:30	9:45	10:00	10:15	10:30	10:45	11:00
0		184	172	70	129	123	117	113	108	106	102	100	100
50		207	201	112	180	159	160	141	133	147	127	119	114
100		161	203	143	233	208	200	175	160	168	145	138	126
150		45	95	96	248	255	237	213	188	188	162	153	136
200		0	5	37	152	274	248	246	207	206	178	166	145
250		0	0	1	88	262	223	232	205	213	190	173	154
						K-values for CO m^2 sec^{-1}							
50		5.91	7.31	7.47	12	9.09	10.7	5.13	6.65	14.7	40.8	B	B
100		1.02	2.32	3.01	13.3	8.38	20	70.0	128	B*	B	B	B
150		2.25	1.41	1.03	6.2	25.5	19.0	85.2	37.6	B	B	B	B
200			0.22	0.87	1.56	11.0	11.0	12.3	12.5	41.2	71.2	B	B
250				0.07	2.93	2.83	5.6	7.73	10.2	9.47	15.2	17.3	B

* B stands for too big to compute.

Table 4. Summary of 15-minute average K_* values

Height, (m)	h(m)	100			150			200			250			300			350		
		Operation			Operation			Operation			Operation			Operation			Operation		
		28	31	33	28	31	33	28	31	33	28	31	33	28	31	33	28	31	33
50		0.07	0.05		0.09	0.05	0.1	0.06	0.04	0.12	0.05	0.05	0.09	0.04	B*	0.07	0.02	B*	0.07
100					0.02	0.02	0.01	0.03	0.03	0.02	0.03	0.04	0.02	0.04	0.08	0.06	0.04	0.08	0.06
150								0.01	0.02	0.01	0.02	0.04	0.01	0.02	0.04	0.06	0.02	0.04	0.03
200								0.00	0.01	0.00	0.00	0.01	0.00	0.01	0.02	0.03	0.01	0.02	0.03
250														0.00	0.01	0.02	0.00	0.01	0.02

Height, (m)	h(m)	400			450			500			550			600			650		
		Operation			Operation			Operation			Operation			Operation			Operation		
		28	31	33	28	31	33	28	31	33	28	31	33	28	31	33	28	31	33
50		B*	B	0.03	B	B	0.06												
100		B	B	0.20	B	B	B												
150		0.03	B	0.09	B	B	B												
200		0.02	0.03	0.03	0.03	B	0.03												
250		0.01	0.02	0.02	0.02	0.02	0.02												

* B stands for too big to compute.

6. DISCUSSION OF RESULTS

To summarize, we have presented four independent estimates of the vertical distribution of the eddy coefficient for dispersion of a passive contaminant from an extensive area source in a convective layer; these estimates were based on the following methods:

(1) A second-order closure prediction

(2) Field data of pollutant concentrations over Los Angeles

(3) Laboratory measurements of particle dispersion

(4) Assumption of equality between momentum and mass transfer coefficients in the free convective limit.

It has been reasoned that if horizontal inhomogeneities are negligible (it was supposed to be so for all estimates) the dispersion process is fully determined by the turbulent structure; hence we should be capable of finding a self-similar solution for the contaminant eddy coefficient provided the contaminant has the surface as its source. Apart from the region close to the inversion base (where the effect of the inversion strength becomes important), we then expect that the methods mentioned above yield consistent results. The various techniques did not agree and we shall now discuss possible reasons for the disagreement and suggest the "best" compromise.

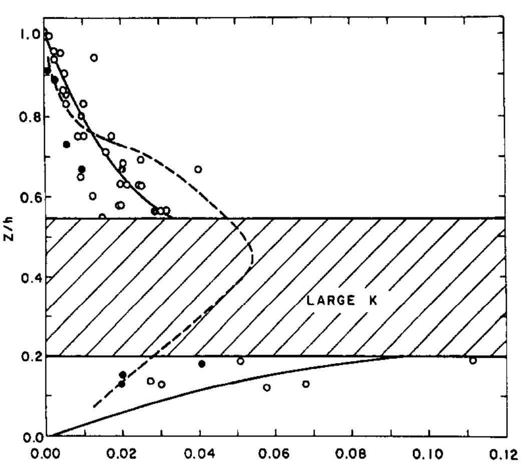

Fig. 3. Distribution of K_* with z/h. Solid lines fit observations. Lower solid line also fits Eq. (9). Dashed line from second-order closure theory.

Compared with the methods (2) and (3), the second-order closure may underestimate K, especially in the central region of the mixed layer. Deardorff (private communication) suggested that a plausible cause for this lies in underestimating the triple correlation term $\overline{\chi'\theta'w'}$ which represents the vertical transport of the quantity $\overline{\chi'\theta'}$. Here, θ is potential temperature; an overbar denotes an average, and a prime denotes deviations from the average. The term $\overline{\chi'\theta'w'}$ effects the transport of $\overline{\chi'\theta'}$ from the surface to the core of the mixed layer, where there is no local production of $\overline{\chi'\theta'}$. In turn, the buoyancy term $g/T_0\ \overline{\chi'\theta'}$ can support the flux $\overline{\chi'w'}$ without a mean gradient $\partial\bar{\chi}/\partial z$. Therefore, $K\chi = -\ \overline{\chi'w'}/\partial\bar{\chi}/\partial z$ can become very large. Examination of the model results indicates that Deardorff's speculation may be correct. Eventually, it should be possible to get better estimates of K by second-order closure methods, once the relevant universal constants are known.

The estimates at Los Angeles also may be subject to systematic errors. In particular, vertical advection and horizontal diffusion were omitted in Eq. (2). The errors produced by this omission can be analyzed (Deardorff, personal communication) by generalizing Eq. (3) to:

$$F_z = \int_z^h \frac{\partial \chi}{\partial z}\,dz - \bar{w}\chi_z + \int_z^h \left[\frac{\partial F_x}{\partial x} + \frac{\partial F_y}{\partial y}\right] dz \quad (12)$$

where $\bar{w}$ is the mean vertical velocity between z and h and F_x and F_y are longitudinal and lateral turbulent fluxes of contaminant, respectively. In the Los Angeles area, subsidence generally exists, as Deardorff points out. Therefore, sinking increases F_z, and thus the omission of the second term on the righthand side of Eq. (12) leads to underestimating of F_z. Numerical estimates on the basis of Fig. 1, and Table 3 suggest that the fluxes may be incorrect by factors of the order 1.5, with similar errors in the mixing coefficients.

The last term in Eq. (12) is presumably small in the center of a large area of polluted air. But, qualitatively, the last term in Eq. (12) is also positive, and its omission would further lead to overestimates of K. Hence, the omission of both vertical advection and lateral diffusion lead to an underestimate of eddy coefficients; but it is difficult to see how these effects could exceed a factor of 2.

The eddy coefficients K_χ estimated from the particle dispersion experiment of Deardorff and Willis are the largest of the estimates presented. It is reasonable to ask a question whether this experiment simulates realistically a gaseous dispersion in the real atmosphere.

Particles imbedded in a fluid at rest perform Brownian motion and their diffusion rate is negligible compared to the diffusion rate of a gas in the same fluid. The lack of molecular diffusion of particles will inhibit the molecular destruction rate of the correlation $\overline{\chi'\theta'}$. This will lead in buoyant flows to intensification of turbulent diffusion in regions where $\overline{\chi'\theta'} > 0$.

A quantitative estimate from the second-order model equations indicates that the eddy coefficient for particles could exceed the gas eddy coefficient for otherwise identical conditions by a factor of 1.5. Hence, the Deardorff–Willis results would have to be scaled down to account for the gas-particle effect.

Estimate (4) is based on measurements of $K_M(= u_*^2/\partial\bar{u}/\partial z)$. The expression in Eq. (9) can be rewritten to yield

$$K_M^* = K_M/w_*h = 0.75\ (z/h)^{4/3}.$$

Although the power exponent 4/3 is correct (common to all eddy coefficients in the free-convection limit) there is no reason to expect an equality between momentum and mass transfer coefficients in a convective surface layer. An approximate equality exists between the heat and mass transfer coefficients. This is due to the similar behavior of the buoyant term $\overline{\chi'\theta'}$ and $\overline{\theta'2}$ close to the surface. The second-order model yields the following quantitative estimates:

$$K_H^* \simeq 2.2\ (z/h)^{4/3}$$
$$K_M^* \simeq 0.75\ (z/h)^{4/3}$$
$$K_\chi^* \simeq 2.0\ (z/h)^{4/3}.$$

Hence, K_H^*/K_M^* is predicted to be on the order of 3, in good agreement with observations. The contaminant eddy coefficient K_χ^* is as expected, close to K_H^*; but K_M^* is significantly less than K_χ^* suggesting again that the Los Angeles estimates of K_χ^* are too small.

It is concluded that K_* is quite possibly a universal function of z/h for a fairly homogeneous ground source under convective conditions. Its magnitude is most likely about twice that given by the solid line in Fig. 3 and somewhat smaller than suggested by the laboratory experiments.

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